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(54) Title of the Invention Dry bleach stable enzyme composition

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Elaboration of specification (no amendment to the contents)

Specification

1. Title of the Invention

Dry bleach stable enzyme composition

2. Claims

1. A water-soluble granulate enzyme composition comprising a core of enzyme material and a protective coating containing an effective amount of alkaline buffer salt having a pH of from approximately 7 to approximately 11, wherein said protective coating surrounds said core and said effective amount of alkaline buffer salt provides improved enzyme stability in the presence of solid peroxyacid bleach granulates.

2. The composition as described in Claim 1, wherein said core is approximately 33 to approximately 90% by weight of said composition.

3. The composition as described in Claim 1 or 2, wherein said protective coating surrounding said core is at least 10% by weight of said composition and wherein said core is approximately 50 to approximately 80% by weight of said composition.

4. The composition as described in Claim 1 or 2, wherein said protective coating contains 50 to 100% alkaline buffer salt by weight of said protective coating.

5. The composition as described in Claim 1 or 2, wherein said protective coating contains 50 to 100% alkaline buffer salt by weight of said protective coating, and wherein when said alkaline buffer salt is present by an amount of approximately 5 to approximately 10% by weight of said composition, the balance of said coating is selected from antioxidants, calcium chloride, and other compatible inorganic salts.

6. The composition as described in Claim 1 or 2, wherein said alkaline buffer salt protective coating has a pH of 8 to 10, and wherein said core, with respect to said core coating, has a weight ratio of 4:1 to 1:1.

7. The composition as described in Claim 1 or 2, wherein antioxidant salts are present in said protective coating by an amount of 1 to 40% by weight of said composition.

8. The composition as described in Claim 7, wherein said antioxidant is present by an amount of 2 to 30% by weight of said composition.

9. The composition as described in Claim 1 or 2, wherein said protective coating is a mixture of alkaline buffer salt and antioxidant, and wherein said mixture has a pH of 8 to 10.

10. The composition as described in Claim 1 or 2, wherein said alkaline buffer salt is selected from the group consisting of potassium bicarbonate, potassium carbonate, tetrapotassium pyrophosphate, tripotassium polyphosphate, sodium bicarbonate and sodium carbonate, and mixtures thereof, and wherein said alkaline buffer salt in said protective coating is present by an amount of 5 to 50% by weight of said composition.

11. The composition as described in Claim 1 or 2, wherein said protective coating contains an antioxidant selected from the group consisting of sodium sulfite, sodium bisulfite and sodium thiosulfate, and mixtures thereof.

12. The composition as described in Claim 1 or 2, wherein said protective coating contains calcium ion as calcium chloride by an amount of 40 to 3000 ppm by weight of said composition.

13. The composition as described in Claim 1 or 2, wherein said composition is surrounded by an overcoating of water-soluble nonionic wax having a melting point of at least approximately 38°C.

14. The composition as described in Claim 1 or 2, wherein said composition contains a nonionic wax overcoat with a melting point of at least 50°C by an amount of approximately 5 to approximately 57% by weight of said composition.

15. The composition as described in Claim 14, wherein said water-soluble nonionic wax overcoat is present by an amount of 10 to 30% by weight of said composition.

16. The composition as described in Claim 14, wherein said water-soluble nonionic wax overcoat is present by an amount of 15 to 25%.

17. The composition as described in Claim

13, wherein said nonionic wax is selected from the group consisting of fatty alcohols, ethoxylated fatty alcohols, higher fatty acids, mono-, di-, and triglycerol esters of fatty acids, e.g., glycerol monostearate, alkylaryl ethoxylates and coconut monoethanolamide, and mixtures thereof.

18. The composition as describe in Claim 17, wherein said nonionic wax is selected from the group consisting of: TAE_{22}, PEG1500-8000, and palmitic acid.

19. The composition as described in Claim 1 or 2, wherein said composition is encapsulated in an alkaline solution-soluble acetate phthalate resin cap.

20. The composition as described in Claim 19, wherein said composition has an overcoat of nonionic wax under said resin.

21. The composition as described in Claim 1 or 2, wherein said composition is encapsulated in a 5 to 57% alkaline solution-soluble acetate phthalate resin by weight of said composition.

22. A water-soluble granulate enzyme composition, characterized by comprising the following processes:

(1) A process in which an enzyme core is coated with 10% to 100% of a protective alkaline buffer salt solution having a pH of 7 to 11, after going through a 15% to 70% solution; and

(2) A process in which said coated core in process 1 is dried in a fluid bed dryer to provide the water-soluble granulate enzyme composition,

(wherein said composition contains from 33% to 90% of said enzyme core and from 5% to 67% of said alkaline buffer salt on a dry weight basis).

23. The method as described in Claim 22, wherein the solution in process 1 also contains an antioxidant to provide 0 to 62% of an antioxidant coating on said water-soluble granulate enzyme composition.

24. The method as described in Claim 22 or 23, wherein said alkaline buffer salt-coated granulate is overcoated with 5% to 57% of nonionic wax after going through an optional step in a fluid bed.

25. The method as described in Claim 22 or 23, wherein said solution in process 1 contains 170 to 300 ppm of calcium as calcium chloride.

26. The method as described in Claim 23 or 24, wherein said core in process 1 is coated with a nonionic waxy material prior to being coated with said alkaline buffer salt.

27. A peroxyacid bleach/enzyme granular mixture composition characterized by comprising an alkaline buffer salt protective coated enzyme granulate and a peroxyacid bleach granulate having a weight ratio of enzyme granulate to bleach granulate of from 1:1 to 1:1500.

28. The composition as described in Claim 27, wherein said ratio is 1:3 to 1:30.

3. Detailed Description of the Invention

Background of the Invention

The present invention relates to an improved granulate enzyme composition and a method for producing the same. The improved

granulate enzyme composition has improved stability when mixed with a peroxyacid bleach granulate

Over the last 20 years, the use of enzymes, especially enzymes of microbial origin, has become more and more common. Enzymes are used in, for example, the starch industry to produce glucose and fructose by means of amylases, amylglucosidases, and glucose isomerases. In the dairy industry, a vast tonnage of rennets is used, and in the detergent industry, proteases are normally used as additives in the washing powders to provide a better effect on proteinaceous stains on the laundry.

U.S. Pat. No. 3,519,570 discloses enzyme-containing detergent compositions and a process for conglutinating enzymes and detergents.

U.S. Pat. No. 3,784,476 discloses a particulate enzyme-containing detergent composition containing a detergent surface-active agent, a water-soluble builder salt, and discrete-shaped inorganic solids containing proteolytic or amylolytic enzymes. It should be noted that this patent does not describe an enzyme core coated with an alkaline buffer salt as disclosed herein.

U.S. Pat. No. 4,106,991 discloses an improved process for forming enzyme granulates through the inclusion of finely divided cellulose fibers into the composition. Optionally, a waxy substance can be used for the granulating agent, or can be used to coat the granulate. This patent claims a granulate composition comprising enzyme, inorganic salts, a granulation binder, and finely divided cellulose fibers as 2 to 40% by weight of the granule.

It is a difficult task to prepare a storage-stable mixture of enzyme containing granulates and dry peroxyacid bleach granulates. In spite of the fact that some commercially available enzyme granulates are advertised as "perborate bleach-stable", they are weak in terms of storage in the presence of strong peroxyacid bleach granulates. It should be noted that peroxyacid bleach granulates are relative newcomers to the dry commercial laundry detergent and bleach market. The term "bleach" as used herein means peroxyacid bleach unless otherwise specified, and the term "peroxyacid bleach powder" and "peroxyacid bleach granulates" are synonymous unless otherwise specified.

Summary of the Invention

The present invention relates to an improved granulate enzyme composition comprising a core of enzyme material and a protective coating comprising alkaline buffer salt. In another aspect, the present invention relates to a method for producing the improved granulate enzyme composition, comprising coating an enzyme core material with an alkaline buffer salt protective coating. The improved granulate enzyme composition is stable when mixed with peroxyacid bleach granulates.

Objective

An objective of the present invention is to provide an improved granulate enzyme composition that can be mixed with a

peroxyacid granulate and stored without rapid loss of enzyme activity. Other objectives are apparent in light of this disclosure.

Detailed Description of the Invention

The present invention relates to an improved water-soluble granulate enzyme composition comprising an enzyme core containing enzymes, fillers and/or binders, and an enzyme-free alkaline buffer salt protective coating that surrounds said core. The alkaline buffer salt protective boating is applied substantially completely around the enzyme core. The alkaline buffer salt protective coating preferably contains 50 to 100% of said alkaline buffer salt. The balance is selected from antioxidants, calcium chloride, and other compatible inorganic salts. The alkaline buffer salt coating has a pH of approximately 7 to approximately 11. The practical level of alkaline buffer salt protective coating is from approximately 10 to approximately 100% by weight of the core, but can be less than 10% or greater than 100%. The key is to surround substantially the core with an effective amount of alkaline buffer salt to protect the enzyme from deactivation when mixed with dry peroxyacid bleach granulates. When factored into the total composition, 10 to 100% becomes 5 to 50% of alkaline buffer salt itself. Some practical ratio levels of enzyme core to coating, overcoating and encapsulating material (defined below) are from 10:1 to 0.5:1, preferably from 4:1 to 1:1, and more preferably approximately 1.5:1.

The percentages as used herein are% by weight of total composition unless otherwise specified.

The improved granulate enzyme composition preferably comprises, based on the percentage by weight of the total composition: 33% to 90%, more preferably approximately 50% to approximately 80, of enzyme core containing enzyme powder and material selected from cellulosic fillers, binders, and inorganic salt fillers, and mixtures thereof; from 5% to 67%, more preferably 10% to 45%, of alkaline buffer salt in the protective coating surrounding said core, said protective coating including 0.5% to 62%, more preferably 2% to 30%, of an antioxidant in the coating surrounding said core; 5% to 57%, more preferably 10% to 30%, of water-soluble nonionic waxy overcoating; and 5% to 57%, more preferably 10% to 30%, of alkaline solution-soluble acetate phthalate resin cap. In the composition of the present invention, the alkaline buffer salt and antioxidant are coated on the enzyme core prior to overcoating with waxy and/or said resin cap.

The improved granulate enzyme composition is prepared preferably with an enzyme powder level of approximately 1 to approximately 20% by weight (0.5 to 10AU/g), more preferably approximately 1 to approximately 10% by weight (0.5 to 5AU/g) of the total composition. The filler and binder in the core can have a ratio from 10:1 to 1:1. A practical level of cellulosic fillers in the total composition can be approximately 2%

to approximately 36%. AU represents Anson units and is a term commonly used in the trade to describe enzyme activity.

As shown in Fig. 1, the stability of the alkaline buffer salt coated granulate enzyme composition of the present invention is further improved by the addition of an antioxidant to the protective coating. The antioxidant is preferably used in the protective coating at a level of 1 to 40% by weight, more preferably 2 to 30% by weight of the total composition. It is preferably applied with the alkaline buffer salt, but may be applied separately. As shown in Fig. 1, the granulate enzyme composition of the present invention is further improved if it has an overcoat of a water-soluble nonionic waxy material. Such an overcoat may be preferably used by an amount of 10% to 30%, and more preferably 15% to 25% of the total composition.

The improved granulate enzyme composition of the present invention may be mixed with other laundry active powders including peroxyacid bleaches, softeners, detergents, etc. Examples of powdered detergent material are disclosed in U.S. Pat. No. 4,404,128. Examples of powdered peroxyacid bleach granulates are disclosed in U.S. Pat. No. 4,473,507.

A preferred mixture is an enzyme-peroxyacid bleach granulate mixture comprising the alkaline buffer salt protective coated enzyme granulate of the present invention and a peroxyacid bleach granulate having a weight ratio of coated enzyme granulates to bleach granulates of from 1:1 to 1:1500, preferably from 1:3 to 1:30. Details of such a preferred mixture are disclosed below.

Alkaline buffer salt

The term "alkaline buffer salt" as used herein means a salt that has a pH of 7 to 11 and provides a comparable pH for the alkaline buffer salt protective coating in the presence of acidic substances for an extended period of time. Thus, the alkaline buffer salt useful in the present invention can be any one of a number of suitable compatible inorganic salts which have a pH of 7 to 11. A pH of 8 to 10 is preferable. The pH of a salt is measured as a 10% aqueous solution of the salt. Some preferred alkaline buffer salts are potassium bicarbonate, potassium carbonate, tetrapotassium pyrophosphate, potassium tripolyphosphate, sodium bicarbonate, and sodium carbonate. Other suitable alkaline buffer salts may be used.

The alkaline buffer salt can compose 100% of the protective coating. However, other compatible materials may be contained, e.g., other inorganic salts, fillers, binders, etc. An aqueous solution of the protective coating ingredients may be used to apply the protective coating to the enzyme core. Preferably, the solution shall contain 170 to 300 ppm calcium as calcium chloride in addition to the other protective coating ingredients.

Antioxidant

The term "antioxidant" as used herein means a substance that resists oxidation or inhibits reaction provided by oxygen or peroxides.

The antioxidant is a stability booster for the alkaline buffer salt coating. The antioxidant increases the stability of the enzyme when used in conjunction with the alkaline buffer salt.

A preferred enzyme granulate protective coating can contain 0.5% to 62%, preferably 1 to 40%, and more preferably 2 to 30% of antioxidant inorganic salt. However, the protective coating must have an effective amount of alkaline buffer salt present therein. Some preferred antioxidant salts are sodium sulfite, sodium bisulfite, and sodium thiosulfate. Other suitable antioxidant salts can also be used.

Method for coating the core with the alkaline buffer salts

The enzyme core used in the present invention may be coated by a number of known apparatuses. Coating in a fluidized bed is preferred. Examples of suitable apparatuses and methods are disclosed in U.S. Pat. No. 3,196,827, No. 3,253,944, and No. 3,117,027.

U.S. Pat. No. 3,117,027 discloses a preferred fluidized bed apparatus that can be used to coat the small enzyme core particles used in the present invention. The fluidized bed will provide granulates substantially uniformly coated with enzymes.

The method for coating the core with the alkaline buffer salts comprises:

(1) preparing an enzyme core granulate having a particle size of 100 to 1600μm, preferably 200 to 800μm, with or without an optional waxy coating. Alternatively, an enzyme core may be provided.

(2) Coating the enzyme core with an effective amount of alkaline buffer salt coating, preferably at a level of approximately 10 to approximately 100% by weight of the core on a dry weight basis. The core should be surrounded by the coating, and the coating should contain an effective amount of alkaline buffer salt.

The protective coating is preferably applied to the enzyme core as a solution with a solid content of 15% to 70% (preferably from 20% to 50%) in a fluidized bed. The temperature range of the solution can be approximately 60 to 82°C (140 to 180°F), and preferably approximately 65 to 77°C (150 to 170 °F). The air temperature of the fluidized bed is 45 to 77°C for the coating/drying operation. The rate of addition of the coating solution and the rate of drying are dependent on the solution concentration, temperature of air, volume, etc.

Calcium present in the coating

The granulate enzyme composition of the present invention can be improved if it contains approximately 40 to 3000 ppm of calcium, calculated as calcium chloride. Calcium can be added to the granulate by using water containing a calcium content of 100 to 500 ppm, preferably 170 to 300 ppm, calculated as calcium chloride in the protective coating solution.

The 24-day storage test results shown in Table 1 show that the Sample B prepared with water of 10 to 16 grain hardness is more stable than Sample A prepared with deionized

water. The Sample B contains approximately 500 ppm to approximately 1000 ppm of added calcium chloride.

[seal]

Table 1

Store for 24days at 100°F(38°C)

Coating	Remaining enzyme Activity
Sample A: $KHCO_3/Na_2SO_3/TAE_{22}$ that has salt applied using deionized water	67%
Sample B: $KHCO_3/Na_2SO_3/TAE_{22}$ that has salt applied using "tap water" at from 10 to 16 grain hardness	85%

The Samples A and B are similar to composition 1 in Table 3, and thus, are identical, except for the coating solution water. TAE_{22} is tallow alcohol condensed with 22 molar of ethylene oxide per mole of alcohol.

Enzyme core

The enzyme core used in the present invention is a smaller granulate than the coated granulate. The core has a particle size of 100 to 1600 μm, preferably approximately 200 to approximately 800 μm, more preferably 300 to 400 μm. A commercially available enzyme core is the T-Granulate" available from NOVO Industri A/S, Bagsvard, Denmark.

A preferred enzyme core granulate and process for forming it are generally disclosed in U.S. Pat. No. 4,106. 991. The process comprises drum granulating an enzyme composition including inorganic salts and a granulation binder, with a liquid phase granulating agent, and finely divided cellulose fibers in an amount of 2 to 40%w/w based on the dry weight of the total composition.

As reported in said U.S. Pat. No. 4,106,991, more specifically, the process for forming enzyme core granulates comprises implementing to a drum granulator 2 to 40% by weight of cellulose in fibrous form, 0 to 10% by weight of a binder as defined herein, enzyme and filler in an amount that generates the intended enzyme activity in the finished granulate, a liquid phase granulating agent consisting of waxy substance as defined herein and/or water in an amount of 5 to 70% by weight, wherein the maximum amount of the waxy substance is 40% by weight and the maximum amount of water is 70% by weight, all percentages refers to the total amount of dry substances, and wherein the implementation sequence of the different materials is arbitrary except that at least a major portion of the granulating agent is implemented after at least a substantial portion of the dry substances is implemented to the granulator, thereafter the granulates are, if necessary, dried in a conventional manner, preferably in a fluid bed.

The granulates prepared this way are reported in said U.S. Pat. No. 4,106,991 to have a higher physical stability and a higher

resistance against abrasion than granulates without cellulose fibers; hence, they are reported to have a very low dust level. They are excellent enzyme core granulates for the present invention.

The cellulose in fibrous form can be sawdust, pure, fibrous cellulose, cotton, or other forms of pure or impure fibrous cellulose.

Several brands of cellulose in fibrous form are commercially available, such as CEPO and ARBOCEL. In a publication from Svenska Tramjolsfabrikerna AB, "Cepo Cellulose Powder", it is stated that, for CepoS/20 cellulose, the approximate minimum fiber length is 500 μm, the approximate average fiber length is 160 μm, the approximate maximum fiber width is 50 μm, and the approximate average fiber width is 30 μm. Moreover, it is stated that CEPO SS/200 cellulose has an approximate maximum fiber length of 150 μm, an approximate average fiber length of 50 μm, an approximate maximum fiber width of 45 μm, and an approximate average fiber width of 25 μm. Cellulose fibers with these dimensions are very well suited for the purpose of the present invention.

The binders used in said method are the binders conventionally used in the field of granulation with a high melting point or with no melting point at all and that of a nonwaxy nature, such as polyvinyl pyrrolidone, dextrin, polyvinyl alcohol, and cellulose derivatives, such as hydroxypropyl cellulose, methyl cellulose, or CMC. Granulates cannot be formed based on the cellulose, enzymes, fillers, and binders without using a granulating agent as defined below.

All enzymes can be granulated by means of said method. Preferably, amylases and proteinases are granulated according to the present invention. Specific examples are ALCALASE (a Bacillus licheniformis proteinase), ESPERASE, and SAVINASE (microbial alcaline proteinases produced according to British Pat. No. 1,243,7849), and TERMAMYL (a Bacillus licheniformis amylase). The enzyme can be introduced into the granulator as a pre-dried milled powder or as a solution, for example, a concentrated enzyme solution prepared by ultrafiltration, reverse osmosis, or evaporation.

The filler is used only for the purpose of adjusting to the intended enzyme activity in the finished granulate. Since the enzyme introduced into the granulator already contains diluent impurities that are considered as fillers, an additional filler is not always needed to standardize the enzymatic activity of the granulate. A preferred filler for the core can be an alkaline buffer salt, or an antioxidant inorganic salt, or mixtures thereof as defined herein.

The granulating agent is water and/or a waxy substance. The granulating agent is always used as a liquid phase in the granulation process. Therefore, the waxy substance if present is either dissolved or dispersed in the water or melted. A "waxy substance" is understood as a substance that possesses all of the following characteristics: (1) the melting point is from 30 to 100°C, preferably

from 40 to 60°C; (2) the substance has a tough and not brittle nature; and (3) the substance has substantial plasticity at room temperature.

Both water and waxy substances are granulating agents, i.e., they are both active during the formation of the granulate cores. The waxy substance stays as a component in the finished granulate core, whereas the majority of the water is removed during the drying process. Thus, in order to refer all amounts to the finished dry granulate cores, all percentages are calculated based on the total dry cores. This means that water, which is one of the granulating agents, is not added to the other components when the percentage of water is calculated, whereas the waxy substance, which is the other core granulating agent, has to be added to the other dry components when the percentage of waxy substance is calculated. Examples of waxy substances are polyglycols, fatty alcohols, ethoxylated fatty alcohols, higher fatty acids, mono-, di- and triglycerolesters of higher fatty acids, e.g., glycerol monostearate, alkylaryl ethoxylates, and coconut monoethanolamide.

An illustrative summary of a method used to prepare an enzyme granulate core is as follows:

1. Prepare dry enzyme powders, fillers, binders, etc.

2. Mix the dry powders of the core composition.

3. Wet the powder mixture with granulating agent, e.g., water or waxy melt.

4. Process the wet powder mixture in process 3 in a granulating apparatus (e.g., rotating knife) to form a granulate core having a desired particle size distribution.

A cylindrical Lodige type mixer FM 130DIZ (U.S. Pat. No. 3,027,102) can be used in the processing of this process. The mixer is equipped with both a plough shaped mixer mounted on a horizontal (axial) rotating shaft and a granulating device, consisting of one or more cross knives mounted on a shaft implemented to the mixer through the cylindrical wall in a direction perpendicular to said horizontal rotating shaft (i.e., radial direction of the cylinder).

5. Dry the wet granulate core in process 4 in a fluidized bed to a dryness level that satisfies both the enzyme stability requirements and free-flowing nature and mechanical strength requirements. Usually, this shall correspond to a water content less than 10, preferably less than 3%, and more preferably a complete dryness. In the case in which the granulating agent is exclusively or principally a waxy substance, only cooling may be required.

6. In an optional sixth step, the granulate in the step 5 can be coated with a waxy or some other compatible substance.

The core is then coated with alkaline buffer salts.

Some preferred enzyme core granulate compositions and ingredient ranges are shown in Table 2.

Table 2

Enzyme core granulate levels

Component	Preferred	Low	High
Proteolytic degrading enzyme	4	0.5	15
Amylase enzyme	1	0	3
Calcium sulfate, $CaCl_2$ Sodium sulfate, NaCl	45	3.0	97.5
Cellulose filler and binder	25	2.0	40
Waxy overcoat (PEG1500)	25	0	40

Such enzyme cores constitute 33 to 90% by weight of the preferred and practical coated compositions of the present invention.

Optional waxy coating substance

A nonionic waxy substance may be applied over the core or over the alkaline buffer salt coated enzyme granulate. The practical levels of waxy "overcoats" are up to 57%, preferably 5 to 30%, and more preferably 15 to 25% by weight of the composition. The term "overcoat" as used herein means that this includes a mixture of an alkaline buffer salt and an antioxidant salt on the alkaline buffer salt coating. Examples of such waxy overcoatings are polyethylene glycols, fatty alcohols, ethoxylated fatty alcohols, higher fatty acids, mono-, di- and triglycerolesters of fatty acids, e.g., glycerol monostearate, alkylaryl ethoxylates, and coconut monoethanolamide. Preferred nonionic waxy substances are TAE_{22} (tallow alcohol condensed with 22 moles of ethylene oxide per mole of alcohol), PEG 1500-8000 (polyethylene glycol with a molecular weight of 1500-8000), and palmitic acid. Other waxy coatings having a melting point of at least 38°C, preferably at least 50°C, can also be used. For example, this waxy coating is melted (50-70°C), and is sprayed onto the granulate in a fluidized bed in which cool air (15-30°C) is applied to solidify the waxy coating.

Figures

Fig. 1 and Fig. 2 show potent graphical illustrations of the improved stability of the alkaline buffer salt coated granulate enzyme compositions of the present invention over some other granulate enzyme compositions. The enzyme granulate compositions 1-5 in Table 3 correspond to the curves 1-5 in Fig. 1 and Fig. 2. The amount of components is reported in Table 3 as percentages of the total granulate enzyme composition. The coating method used to prepare compositions 1-3 and 5 are set forth in Example II.

Table 3

Enzyme granulate compositions

	1	2	3	4	5
Curve	%	Wt%	Wt%	Wt%	Wt%
Coating (T-Granulate)	61.5	61.5	80	100	80
Potassium bicarbonate	15.4	18.5	20	-	-
Sodium bisulfite	3.1	-	-	-	-

TAE_{22}	20.0	20.0	-	-	20

Four grams of each composition (1-5) in Table 3 were mixed with 20 grams of the peroxyacid bleach composition in Example III. For Fig. 1, the stability tests were performed at approximately 100°F (38°C) and ambient humidity. For Fig. 2, the stability tests were performed at approximately 80°F (27°C) and 15% relative humidity. In both tests the Enzyme Stability (ES) curve 1 is the best one. Thus, the composition 1 in Table 3 represents a potent embodiment comprising an alkaline buffer salt/antioxidant coated granulate enzyme composition with an overcoat of TAE_{22} in the presence of a peroxyacid bleach as set forth in Example II. Enzyme Stability (ES) curve 2 shown in Fig. 1 and Fig. 2 is the next best one. It should be noted that the composition 2 in Table 3 is identical to the composition 1, except that this has no antioxidant. ES curve 3 is identical to "2" that has no overcoat, TAE_{22}.

ES curve 4 is a prior art overcoat T-Granulate, and ES curve 5 is a prior art T-Granulate with additional TAE_{22} overcoating.

Similar potent stability results were obtained at a lower temperature (27°C) and 15% relative humidity as shown in Fig. 2.

Example I

A preferred enzyme core can be produced by means of said method using the following components:

Component	Wt%
Proteolytic enzyme	4
Amylase enzyme	1
Calcium sulfate, $CaCl_2$	45
Sodium sulfate, NaCl	
Cellulose filler[1)]	20
Binder[2)] (polyvinyl pyrrolidone)	5
Waxy overcoat (PEG1500)	25

1) Cellulose powder-CEPO S20

2) Selected from polyvinyl pyrrolidone, dextrin, polyvinyl alcohols, and cellulose derivatives

Example II

A 6 inch (about 15.2 cm)-wurster fluidized bed coating unit with a capacity of about 1 liter was used. The preparation method of the coated enzyme is as follows: 800 g of enzyme T-Granulate is added to the fluid bed dryer. To this, 1,000 g of 70°C aqueous solution containing 200 g of potassium bicarbonate and 40 g of sodium sulfite are sprayed on. The coated granulate enzyme composition is then dried at a fluid bed temperature of 75°C such that this contains less than 0.5% of water. The coated granulate enzyme is then removed from the fluid bed dryer and weighed to confirm the coating level.

Subsequently, 800 g of the alkaline buffer salt/antioxidant salt-coated granulate enzyme was placed back into the fluid bed dryer. To this 200 g of TAE_{22} was splayed on at 55°C and allowed to cool in the dryer that have an air temperature of 20°C.

Final weight%:

Enzyme T-granulate 61.54%

core	
Coating:	
Potassium bicarbonate	18.46
15. 38)	
Sodium sulfite 3. 08)	
TAE_{22} overcoating	20.00
Total	100.00%

The ratio of enzyme core to coating is approximately 3.3 to 1. The PH of the coating is 8.5.

The coated enzyme from Example II is mixed with the dry peroxyacid bleach composition as described below in Example III. The stability was tested against the stability of uncoated T-granulate, a TAE_{22} coated T-Granulate, a potassium bicarbonate coated T-Granulate, and a potassium bicarbonate + TAE_{22} coated T-Granulate. These compositions are shown in Table 3, and the stability results are shown in Fig. 1 and Fig. 2.

Example III

The coated enzyme granulates similar to that described in Example II are dry mixed with peroxyacid bleach granulates in the following proportions.

	Weight %	Grams
Peroxyacid bleach granulate		
Diperoxydodecanedioic acid	20.75	
Dodecanedioic acid	1.85	
Boric acid	22.75	
Na_2SO_4	28.06	
Acid sodium pyrophosphate	5.00	
$C_{13}LAS$	4.50	
	83	20

Coated enzyme granulate of Example III

Enzyme core*	10.5	
$KHCO_3$	2.6	
Na_2SO_3	0.5	
TAE_{22}	3.4	
	17	4
	100	24

*Enzyme core is Novo T-Granulate having 2.0 Au/g of protease activity. Its approximate composition is shown in Example I.

The method used to prepare the peroxyacid bleach granulate in Example III is disclosed in U.S. Pat. No. 4,497,757.

The peroxyacid bleach/enzyme granule mixture composition from Example III comprising the alkaline buffer salt protective coated enzyme granulate and a peroxyacid bleach granulate having a ratio of 1:5 was storage stable for more than 10 weeks at 38°C. Thus, the present invention provides an improved enzyme granulate that is storage stable with a peroxyacid bleach granulate, enabling them to be used together in a detergent or laundry additive product for combined bleaching and stain removal performance.

4. Brief Description of the Drawings

Fig. 1 and Fig. 2 are graphs illustrating the stability of compositions of the present invention vs. various coated and uncoated enzyme granulates in the presence of a dry peroxyacid bleach granulate composition.

Applicant representativeKazuo Sato

1/2

Fig. 1

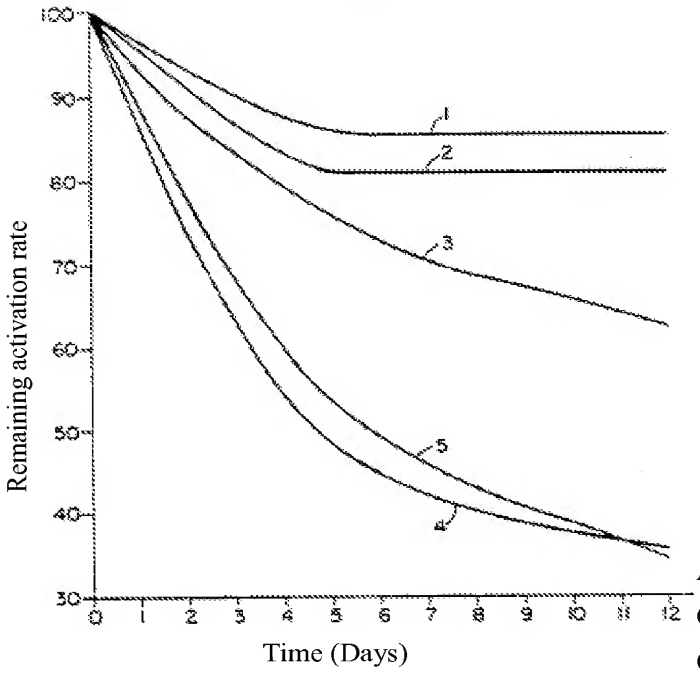

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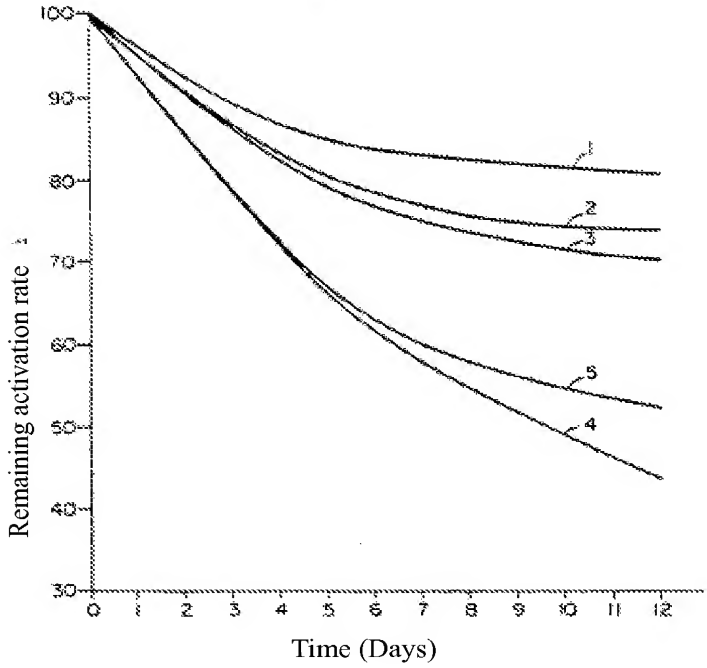
Fig. 2



Amendment (Form)

October 23, 1986

Commissioner of JPO, Akio Kuroda

1. Indication of the Case

Patent Application No. S61-151358

2. Title of the Invention

Dry bleach stable enzyme composition

3. Person who makes amendment

Relation to the case: Patent applicant

The Procter & Gamble Company

4. Representative (Postal code 100)

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Representative

6428 Kazuo Sato, patent agent

5. Date of order for amendment
September 3, 1986
(Mailing date: September 30, 1986
6. Object for amendment
Column for patent applicant of the application, power of attorney, specification, drawings.
7. Contents of amendment
(1) As described in separate sheet
(2) Engrossment of specification and drawings (no amendment to the contents)